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Transmission of visible light through mortars using fluorite as a fine aggregate

Transmisión de luz visible a través de morteros con fluorita como agregado fino

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ABSTRACT: This article presents the results of an evaluation of the optical properties of white and gray Portland cement mortars within the visible light spectrum. Eight controllable variables were studied: size of aggregate, type of cement, water-cement ratio, type of aggregate, cement-aggregate ratio, thickness of mortar, percentage of air entrained and percentage of optical fiber. A mathematical model was used to analyze the behavior of a light beam as it passes across a mortar. Three properties were measured: reflection, absorption and transmittance. A spectrophotometer and two sources of light were used. Of the eight controllable variables, thickness, aggregate size and cement type were those with the greatest impact on transmittance.

RESUMEN: En este artículo se presentan los resultados de la evaluación de las propiedades ópticas, en el intervalo de luz visible, en morteros de cemento Portland blanco y gris a partir de ocho variables controlables: tamaño del agregado, tipo de cemento, relación agua-cemento, tipo de agregado, relación cemento-agregado, espesor del mortero, porcentaje de aire incluido y porcentaje de fibra óptica. Se definió la modelación matemática que permitió identificar el comportamiento de un haz de luz a medida que atraviesa los morteros. Se midieron tres propiedades: reflexión, absorción y transmitancia. Se utilizó un espectrofotómetro y dos fuentes de luz. De las ocho variables controlables el espesor, el tamaño del agregado y el tipo de cemento son las de mayor incidencia sobre la transmitancia.

1. Introduction

The use of artificial energy in buildings generates a high consumption of the natural resources used to produce it, which has the consequence of increasing the scarcity of primary materials that are mostly non-renewable [1]. One way of reducing the use of these materials is to use natural light. However, the existing options for materials permitting light to pass complement, but do not substitute for, conventional construction materials. For this reason, one must decide between materials that increase resistance and structural rigidity, and translucent materials. In many cases, this significantly limits the area available for materials that make it possible to illuminate the interiors of residential or industrial buildings using natural light. The most-used construction material is concrete, the properties of which do not include translucence [2]. Clearly if it were possible to give concrete this property, this would help reduce the use of artificial light. At a global level, three

lines of research have been undertaken in regard to this issue: the first is based on replacing Portland cement with translucent polymer cement; the other two have focused on complementing or substituting the aggregate with materials that permit an electromagnetic wave to be transmitted within the visible light spectrum [3, 4]. The best results have been observed in mixtures composed of thousands of optical fibers aligned in the same direction as the incident light beam, and in mixtures in which the traditional cement is replaced by a polymer with better optical properties (of up to 80% transmittance) [5-13]. When a light beam impacts a material, one or more of three physical phenomena occur: reflection, absorption or transmittance. The first of these depends on the value of the refractive index, which describes the relationship of the speed of light in a vacuum with the speed of light in a substance. This is directly proportional to the losses due to reflection. The other two phenomena occur as a function of the thickness of the sample; the extinction coefficient, which depends on the composition of the material; and the concentration of the absorbent [14]. The type of aggregate used in preparing the mortars was chosen based on the following characteristics: it possessed translucency and it had the least negative reaction with the cement. Calcium fluoride, known as fluorite, meets these criteria in its pure state. Because it is an ionic solid, the conduction band and valence band are

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separated by a forbidden gap of relatively high energy (> 3.5 Electron volts [eV]), which allows the energy produced by the photons in wavelengths in the visible spectrum (of 1.8 eV to 3.1 eV) to avoid interacting with the valence electrons of a fluorite solid, thus permitting the light beam to cross it [14, 15]. If fluorite is used as an aggregate, it is unlikely to have a reaction to the cement alkali or any other destabilizing reaction, since in its pure state, it lacks silica, clays, sulfides or sulfates that could cause detrimental volumetric changes within the mortar or concrete [16]. Finally, calcium fluoride is a natural and abundant material [15] that could reduce the costs of manufacturing the mixtures. Some reports on translucent concrete are limited to reporting the evaluation of its percent transmittance and its mechanical behavior under compressive stress, and do not include data regarding absorption, reflection, concentration of absorbent materials, the extinction coefficient or the thickness of the sample. This makes it difficult to directly compare one material to another based on optical properties. Unlike in the literature found, until now, transmittance was not the only thing evaluated in this study; a characterization was also performed of reflection and absorption properties of the mortars that were used. Loss of energy in the light beam as it crossed the material was also determined, which made it possible to understand its behavior from the moment that it passed from the air to the mortar, then traveled through the interior of the mortar, until the point when it finished crossing and exited into the air.

2. Theoretic supplement

In searching for preexisting information regarding the behavior of a solid when it interacts with a visible light beam, it was found that the principal restriction is that the current model, developed by Bouguer-Lambert-Beer, is valid principally for homogenous and isotropic materials [14]. This condition is not met by the mortars evaluated. In order to develop an approximate explanation of the phenomenon being evaluated, it was necessary to develop a theoretic supplement using the existing model. The mathematical supplement was used as a theoretical argument in order to explain the phenomena observed during experimentation. It will be necessary in future research to validate this model. Through the review of the literature and the theoretic supplement, it was possible to determine the variables to be controlled during the experimental process [17]. Eight variables were accounted for: size of aggregate, type of cement (according to color), water-cement ratio, percentage of an additive that contains air, percentage of optical fiber (without any particular arrangement), type of aggregate, cement-aggregate ratio and thickness of the mortar sample. Each one influences in some way the behavior of the optical properties, since these variables are related to the properties involved in an optical characterization of each material, which are: refractive index, extinction coefficient, concentration of the absorbent and thickness [14]. This article shows the results for the mortar thicknesses that showed the transmittance values and variables accounted for with the highest degree of impact on the dependent

variables. When a light beam with an intensity of I_o impacts a solid, the following can occur (Figure 1):

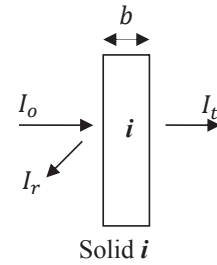


Figure 1 Sequence of light beam in solid *i*

The light beam can be reflected from each one of the sides that it crosses with an intensity of I_r , can undergo absorption in the interior and, finally, can be transmitted with an intensity of I_t . The percentage absorbed depends on the thickness, b , and the characteristics of the mass that it is passing through. The Bouguer-Lambert-Beer law [14, 18] describes the trajectory shown in Figure 1 and defines the intensity of the light beam transmitted using Eq. (1).

$$I_t = I_o(1 - R)^2 e^{-kcb} \quad (1)$$

I_t is the intensity of the light beam transmitted, I_o is the incident light beam, R is the percentage of loss due to reflection, b is the thickness of the sample, and the product kc is known as *Absorptivity*, where k is the extinction coefficient, and c is the concentration of the absorbent [18]. In the case of mortars that are constituted by two mediums, cement paste and aggregate, which are alternately linked several times, the following occurs when they interact with a light beam (Figure 2):

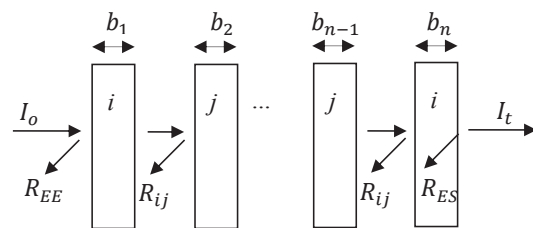


Figure 2 Sequence of a light beam in the materials *i* and *j*

Transmittance results from the loss of the intensity of the incident light beam, due to two external reflections, R_{EE} y R_{ES} (which are produced when the light beam enters and leaves the solid); the internal reflections, R_{ij} (produced at the borders between the aggregate and the paste); and the absorption that occurs during the alternating passage through each material. From a mathematical procedure similar to that developed by Bouguer-Lambert-Beer, it is possible to determine a mathematical expression based on times that the light beam travels through *i* and *j* materials (Figure 2). The mathematical description of the movement of the light beam is expressed in Eq. (2).

$$I_t = I_o(1 - R_{EE})(1 - R_{ES})(1 - R_{ij})^{n-1} (e^{-(kcb)_i})^x (e^{-(kcb)_j})^y \quad (2)$$

Where:

n : Number of times that the beam crosses the cement paste and the aggregate.

x, y : Exponents that depend on n , as follows:

$$\text{If } n \text{ is even } \left\{ x = y = \frac{n}{2} \right\}$$

$$\text{If } n \text{ is odd } \left\{ x = \frac{n+1}{2}; y = \frac{n-1}{2} \right\}$$

In Eq. (2), I_t is a function of six factors. The first of these, I_o , is the intensity of the incident light beam; the second and the third, $(1-R_{EE})(1-R_{ES})$, refer to the losses due to external reflection, that is to say, when the light beam reflects off the surface of the solid at the point it is entering and when the light beam reflects off the surface of the solid it is leaving; the fourth, $(1-R_{ij})^{n-1}$, indicates the losses due to internal reflection, that is to say, when the light beam travels inside the material and hits the paste-aggregate borders; and finally, the fifth and sixth, represented by $(e^{-(kcb)_i})^x (e^{-(kcb)_j})^y$, refer to the losses due to absorption causes by the paste and by the fluorite, respectively. If in Eq. (2) we assume that n is equal to 1, it means that i and j materials are the same, therefore the two external reflections (R_{EE} and R_{ES}) are equal and the sum of x and y would be 1. By replacing these values in Eq. (2) we obtain the Eq. (1). Proving that the starting point corresponds to the initially raised by Bouguer-Lambert-Beer.

3. Experimentation

In each sample evaluated, three optical properties [dependent variables] were measured through the implementation of a fractional factorial design [17]: Reflection, Absorption and Transmittance. Each one of the controllable variables was assigned a level, taking into consideration the availability of material and specifications commonly used in the preparation of mortars. In Table 1, the values for each variable are identified.

Table 1 Values of the controllable variables

Controllable variables	Levels	
	Low	High
Size of aggregate (average in mm)	1.015	3.56
Type of cement	Gray	White
Water-cement ratio	0.50	0.60
% air (by weight)	0	0.50
% optical fiber (by weight)	0	0.05
Type of aggregate	Fluorite	Glass
Cement-aggregate ratio	1:2.75-1:3-1:3.75-1:4	
Thickness of the sample (mm)	5, 10, 20, 50	

The materials used to prepare the mortars were:

Cement: The type of cement was Portland Type III, which is commonly used in the construction industry. It is available in the region, and is manufactured both in gray and in white.

Fluorite: Colombian fluorite from a mine located in the Zaragoza municipality of the Antioquia department.

Air entrainer: Liquid additive of an amber color based on neutralized resins, which complies with the standard specification ASTM C260/C260M - 10^a.

Optical fiber: The optical fiber used in the tests was a material frequently used by Medellín's public utilities company, EPM (Empresas Públicas de Medellín) for data and imaging transmissions. Its diameter was 0.12 mm.

Glass: The glass was a commercial brand classified as transparent glass pane. It was used as a comparative reference to the mortars prepared with fluorite.

Using the primary materials specified and the different levels of the controllable variables, mortars were prepared following the ASTM C109/C109M-08 standard specification. Later, they were placed one by one in the experimental setup (Figure 3), in order to measure transmittance, reflectiveness, and absorbance.

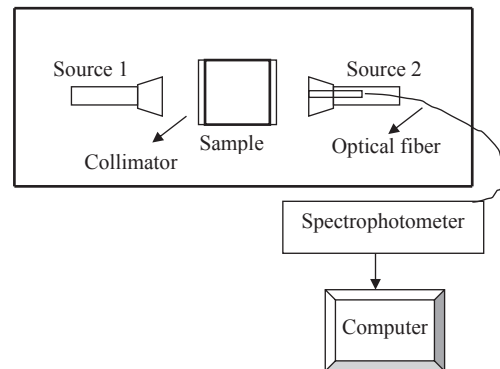


Figure 3 Experimental setup

Sources 1 and 2 in Figure 3 were lanterns with high-efficiency white-light bulbs in the LED (Light-Emitting Diode) matrix, uniform illumination for their full range, and wavelengths that produced a white-light beam within the visible spectrum. A detector was installed in source 2, which, depending on the source that was lit up, detected transmittance or reflectiveness. The information compiled by the detector (optical fiber cable) was sent to an Ocean Optics 2000 spectrophotometer. For each sample, reference spectra were taken 15 minutes after the light source was turned on, with a continuous emission of wavelengths between 350 nm-750 nm. With the goal of preventing the equipment from becoming saturated, an effort was always made to keep the number of counts within the maximum recommended by the manufacturer (3500 counts). At power on source 1 or source 2, the spectrophotometer records a curve within the visible spectrum. The area under

the curve corresponds to the intensity of reflected light [Source 2 on] and the light intensity transmitted [Source 1 on]. From the knowledge of these two intensities and based on Eq. (3) [14] the intensity of absorbed light was calculated, considering that the sum of the three intensities must be 100%.

$$I_o = I_t + I_r + I_a \quad (3)$$

Knowing the intensity of reflected light and based on Fresnel ratio [Eq. (4)] [18] the refractive index (n_2) was determined.

$$R = \left(\frac{n_1 - n_2}{n_1 + n_2} \right)^2 \quad (4)$$

R is the reflectance, which was obtained from the relationship between I_r and I_o . n_1 corresponds to the refractive index of air, that for this investigation an approximate value of 1 was used.

4. Results and discussion

As points of reference, from Eq. (4), the refractive index was calculated independently for the fluorite and the cement pastes, as seen in Figure 4.

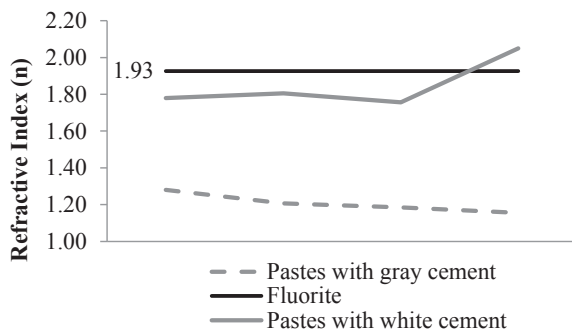


Figure 4 Refractive index in pastes

The horizontal line represents the refractive index value for the fluorite, which on average, was greater than that seen for the pastes of both white cement (continuous line) and gray cement (dotted line). This indicates that on average, of the three materials, the one with the highest value for the intensity of reflected light is fluorite, followed by white cement paste and lastly by gray cement paste. In some white cement pastes, an increased refractive index was observed. This is shown in Figure 4. However, error and standard deviation of the measurements overlap, indicating that variations in the refractive index are not statistically significant. These differences can be related to changes in the degree of homogenization of pastes. Therefore, it was assumed that the average refractive indices of reference are: 1.21, 1.93 and 1.85 for gray cement paste, fluorite and white cement paste respectively. Figure 5 shows the variation in the percentage of

transmittance [y-axis] for fluorite alone, in different thicknesses [x-axis].

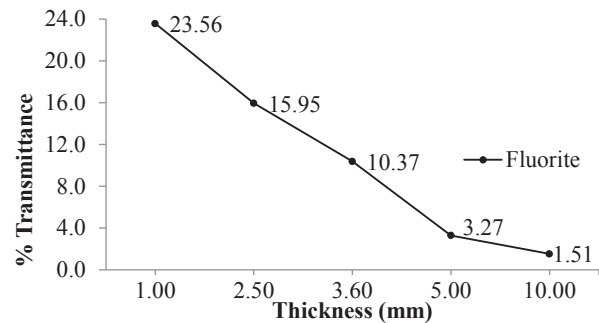


Figure 5 Percent transmittance in fluorite at different thicknesses

For example, for thicknesses of 1 mm and 3.6 mm, which were the most used in the preparation of the mortars, the percentages of light transmitted by the fluorite, as seen in Figure 5, were 23.56% and 10.37%, respectively. Initially, one might think that the ideal would be to use aggregates of a smaller size within the mortars in order to achieve better transmittance, as shown in Figure 5, but as explained below, this option produces an inverse result. For thickness mortars lower or equal to 1 cm, it was possible begin to get results transmittance. The effect of cement type and aggregate size on the percentage of light reflected in the mortars with 1 cm of thickness is seen in Figure 6.

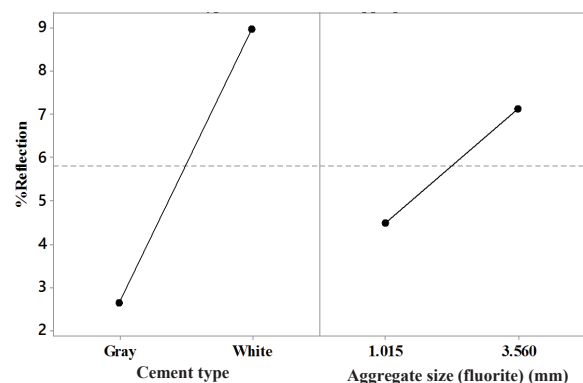


Figure 6 Effect of cement type and aggregate size on reflection

White cement mixed with a larger aggregate size (3.56 mm) produced the greatest losses due to reflection. The y-axis shows the value of the intensity of reflected light and the x-axis shows the types of cement that were used, according to their color, and the average size of the aggregates in mm. Since the white cement paste has a higher refractive index than the gray cement, as seen in Figure 4, the mortars with this cement have a greater intensity of reflected light. When the aggregate size is increased inside the mortars, this increases the possibility that the light beam impacting the mortar can hit fluorite and, since the fluorite has a higher refractive index than the pastes, this would explain

why higher reflection would occur with larger aggregate sizes. The effect of cement type and aggregate size on the intensity of light absorption is seen in Figure 7.

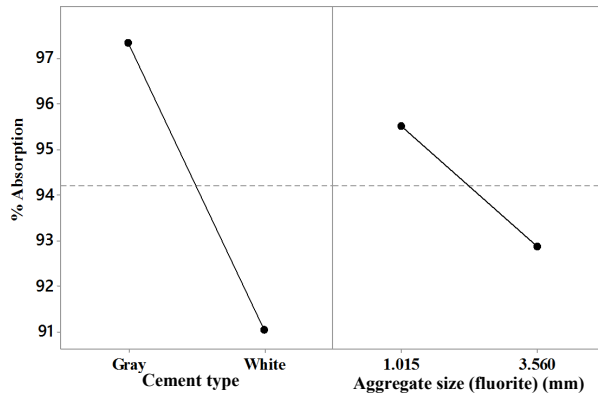


Figure 7 Effect of cement type and aggregate size on absorption

The results regarding absorption, calculated from Eq. (3), were contrary to those for reflection. The mortars with white cement and with the larger aggregate sizes produced the lowest losses due to absorption. The differences in the quantities of light absorbed, when considering the color of the cement, are due to a difference in the atomic structure of bands in the white cement and the gray cement. White cement should have a forbidden energy gap that is greater than that of gray cement, which allows a higher flow of visible light to cross it. The presence of iron oxides in gray cement affects its atomic structure, increasing its capacity to absorb visible light. The decrease in the intensity with which light is absorbed in the mortars when the size of the aggregate is bigger can be explained by the theoretic supplement.

Eq. (2) shows that expression $(1-R_{ij})^{n-1}$ increases as the number of paste-aggregate borders increases, which leads to a decrease in the intensity of the light as it passes through the material, meaning that the amount of light absorbed by the mortar increases. Since the intensity of the incident light beam is equal to the sum of the reflected, absorbed and transmitted light beam, based on the above results it is clear that with white cement and a larger aggregate size, the intensity of the transmitted light beam increases. Later in the experiment, the fluorite aggregate was exchanged for glass and the effect that an aggregate with greater translucency than fluorite had on the quantity of light transmitted through the mortars with white cement and 1cm thick was examined. The y-axis in Figure 8 shows the intensity of the light beam transmitted and the x-axis shows the type of material.

It can be observed that transmittance increases nearly 15 times when fluorite is exchanged for glass. In terms of reflection, the glass and the fluorite, by themselves, have refractive indexes of 1.5 [18] and 1.93 (see Figure 4 for fluorite), respectively. This indicates that fluorite should lose more to reflection. For thicknesses of an average of 2.5 mm, the percentage of the light beam transmitted through

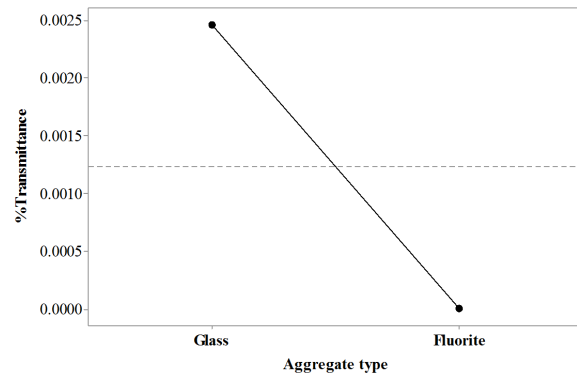


Figure 8 Effect of aggregate type on transmittance

fluorite and glass is 15.95% (Figure 5) and 92% [18], respectively. It should follow, then, that if glass is put into the mortars as an aggregate, the quantity of light transmitted should increase. Finally, the amount of fluorite in the mortars with white cement and 1cm thick was increased, thus modifying the cement-aggregate ratio (1:2.75, 1:3 and 1:4), in order to see what effect this had on the quantity of light transmitted. Figure 9 shows the peak for the cement/fine-aggregate (C:FA) dosage at point 4 (cement-aggregate ratio 1:3) of the x-axis. The y-axis shows the quantity of light transmitted.

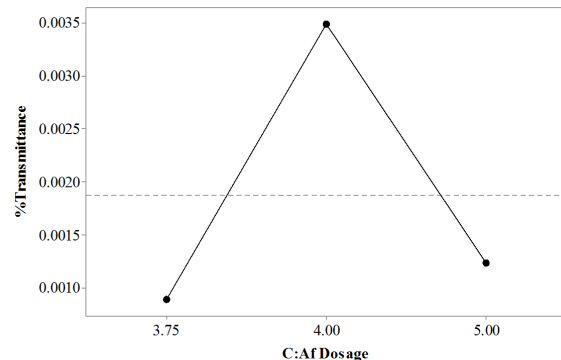


Figure 9 Effect of the cement/fine-aggregate (C:FA) ratio on transmittance

There is an optimal value for the quantity of fluorite that the mortar should have, in which the product of the losses due to internal reflection, represented by the expression $(1-R_{ij})^{n-1}$ and due to absorption, represented by the expressions $(e^{-(kcb)_i})^x (e^{-(kcb)_j})^y$ [see Eq. (2)], produces the maximum transmittance. A lateral displacement from this point can produce smaller losses from reflection but higher losses due to absorption, or vice versa. The decrease in the percentage of absorption when the cement-aggregate ratio is increased is due to a decrease in the width of the space between the aggregates, which reduces the volume of the paste. If the cement-aggregate ratio is increased to higher than the optimal point, despite the fact that the volume of the paste decreases, the quantity of internal paste-aggregate borders increases, which generates greater losses due to internal reflection. Figure 10 shows that transmittance is

also affected by the water-cement (w/c) ratio and by the incorporation of air into the mixture.

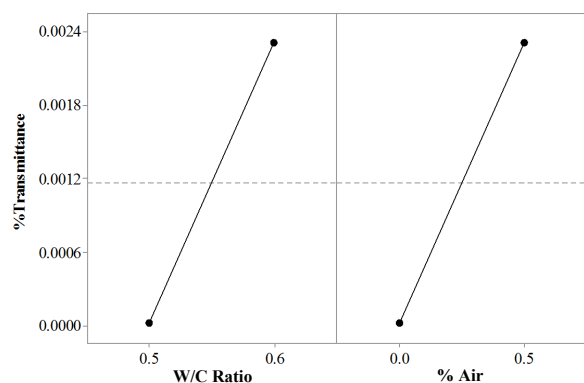


Figure 10 Effects of the water/cement (w/c) ratio and the % of air on transmittance

Higher transmittance is caused by a decrease in the net volume of paste per unit of the volume of the mortar [16] when this is replaced by air, a material with a lower refractive index than the paste [18]. Similarly, when the water-cement (w/c) ratio is increased, the concentration of the absorbent (cement) decreases, thus increasing transmittance. However, the expected effect is not produced when optical fiber is added (see Figure 11), since, as observed, when the fiber is added transmittance decreases. This could perhaps be because the percentage added was not sufficient or because the fibers did not align in a way that facilitated the transmission of the light beam, or for both reasons.

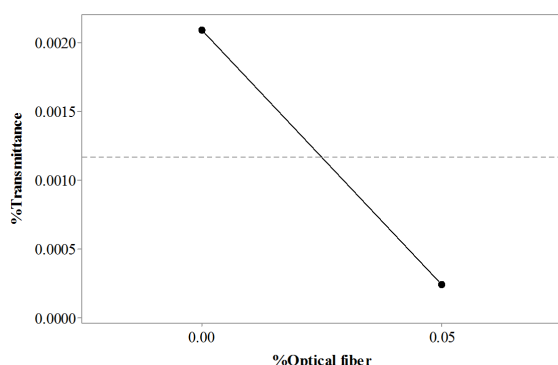


Figure 11 Effect of the % of optical fiber on transmittance

Within the literature reviewed for this study, no mathematical tool was found that allowed the behavior of a light beam within a composite material to be understood. The Bouguer-Lambert-Beer law describes the trajectory of a light beam within homogenous and isotropic materials, characteristics that are not present in a material such as mortar or concrete. The theoretic supplement that was developed in this investigation made it possible to understand how inside this type of composite, the borders between each material exponentially impact the loss of energy in a light beam that is crossing the material. This parameter should be taken into consideration

when choosing the doses for the mortar mixtures, and equilibrium between aggregate size and quantity should be sought. Controlling these variables ultimately makes it possible to control the quantity of visible light transmitted into the interior of a building. Another element that was not found in the literature regarding this subject was a measurement of the optical properties of the materials used. Neither the quantity of light reflected and absorbed, nor the extinction coefficient nor the refractive index was reported. These parameters make it possible to compare one material with another, from an optical perspective. Measuring these parameters for the materials used in this investigation creates a precedent that makes it possible to optically compare these materials to others and to determine the best behavior, based on specific needs, for their use in the desired application. In this study, various variables that commonly affect the behavior of mortars were analyzed from an optical perspective, which made it possible to determine the factors that most greatly impact a mortar's or a concrete's ability to transmit visible light. Until now, the research that has been performed at a global level has been limited to reporting the effect of these factors on the mechanical properties. Using the results of the present investigation, a person responsible for designing mortar or concrete mixes can identify the variables that most influence light transmittance. Future investigations should focus on increasing mortar or concrete thickness in such a way that translucent concrete can be used structurally. Another possibility would be to modify the atomic structure of the cement, in order to reduce light loss through reflection or absorption.

5. Conclusions

Variations in the properties of reflection, absorption and transmittance in the mortars evaluated depended on three principal factors: the thickness, the size of the aggregate and the type of cement. In order to transmit visible light in a mortar, large aggregate sizes should be used for fluorite with white cement, as long as this guarantees an optimal cement-aggregate ratio. The mortars that presented the best behavior in regards to visible light transmission were those that presented the least optical absorption, even if they had higher losses due to reflection.

The theoretical complement made it possible to determine that there must be a balance between the absorption and internal reflection in mortars to achieve higher transmittance. That is, between the thicknesses of the paste surrounding the aggregates and the number of borders paste-aggregate. This parameter must be taken into account when dosed mortar mixtures. It should seek a balance between size and quantity of aggregates.

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